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UV ABSORPTION BY HYDROGEN IN ALKALI METAL. (U)
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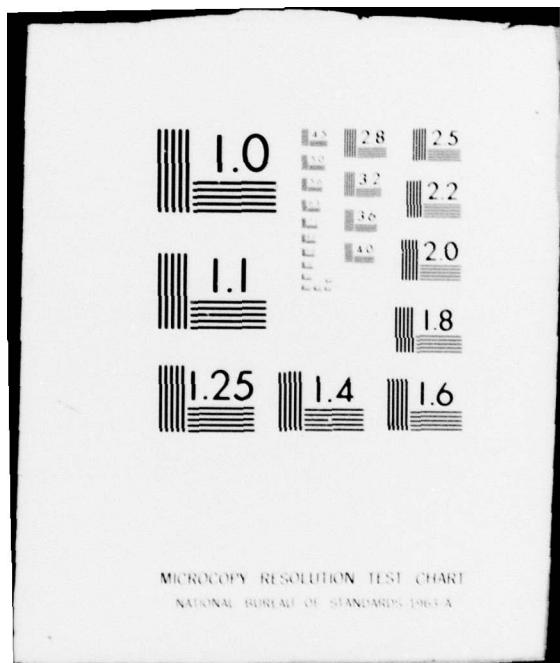
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Half Yearly Report on ONR N 00014-75-C-0918

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The past half year has seen major developments in three areas on which this project focusses. One project - the study of atomic H in alkali metals has been successfully completed and closed out. The student performing that work - Dr. R. P. Layton - graduated with his Ph.D. from the Physics Department on November 12, 1976 for the research supported by ONR. He is now working for the Navy on high pressure studies at Bethesda Naval Hospital. He has been replaced in the research program by Mr. J. A. Cunningham who is working with Dr. J. Erskine on the surface studies projects. The three areas of activities will be covered separately in what follows:

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1. Atomic H in Alkali Metals. This project has been completed successfully and with interesting results. The H source was perfected, as projected in the proposal, to allow fabrication of highly reproducible random alloys containing H impurities. We have discovered that H enters alkali metals interstitially much as is the case for bcc transition metals. The optical spectrum turns out to provide an unusual signature of this interstitial location. As shown in Figs. 1 and 2 for H and O impurities in Cs, these interstitials give a novel sharp peak at ~ 11.6 eV. This peak is entirely lacking from other (substitutional) impurity spectra studied in related projects funded by NSF (e.g. Kr, Xe, F, Cl, Br, I impurities). To verify this deduction we have also completed transport measurements on the thin film alloys. These reveal properties (small residual resistances) that confirm interstitial location of both H and O. The sharp optical peak in

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Cs arises from the way Cs O_{23} excitations at about 12 eV are perturbed by the embedded interstitial neighbors. We expect similar results for the Rb N_{23} edge, the K M_{23} edge, and so forth but these lie beyond the 12 eV cutoff of our present substrates. In measurements of H and O optical properties in Cs, Rb, K and Na up to 12 eV the spectra exhibit features that point to the strong mixing of H and O orbitals with the electron gas, thereby probably making detailed magnetic studies uninformative. They point to wavefunctions near the H in Na, for example, that probably resemble those in the valence band of NaH. Unusual plasmon effects are observed for $\hbar\omega < 12$ eV in Na alloys. The details are provided in Dr. Layton's thesis (copy enclosed) and will be written up in manuscript form for publication in due course.

2. Optical Spectroscopy of Adsorbed Atoms. This very difficult project has yielded novel and informative spectra that raise puzzling questions and present new challenges. Fig. 3 shows a 4.5 monolayer film of Xe on Au, observed in reflection at 7° angle of incidence with the light polarized perpendicular to the surface. The Xe exciton effect, as modified by the surface, are observed at very high signal-to-noise ratio by the present system, which was built and developed with ONR support. This equipment is now functioning with its designed performance level. Spectra about half the amplitude of Fig. 3 are observed for films of 1.5 monolayer thickness and only weak Xe effects are observed from submonolayer films. We have concluded that the initial layer has properties very different from those of subsequent layers. Similar effects are observed for Xe on other metal surfaces. Fig. 4 shows by way of example spectra for 4, 2 and 1 monolayer coverages of Xe on clean Ti. The "exciton" peaks have amplitudes approximately proportional to $n-1$ for n layer coverages. This shows clearly that the first monolayer has a strongly modified excitation

see page 5

spectrum.

These are the explicit surface effects our experiments were intended to investigate. The excited state of the surface layer couples strongly to the conduction electrons of the metal and the sharp spectra of free Xe atoms are heavily broadened at the surface by the excitation of added conduction electron-hole pairs. In the present case the strength of these effects has surprised us. The broadening due to quasiparticle-pair creation for Xe on, for example, an Au clean surface is apparently much larger than that for an Xe atom in bulk alkali as revealed in work under NSF support in our laboratories.

The provocative questions presented by the observed major modification of the surface optical spectrum can only be resolved by studies that reveal the true spectral response of surface layers. We are engaging four separate paths to this goal, which will be detailed in what follows.

1. The use of substrates that couple more weakly to the impurity excited state; 2. The investigation of adsorbed atoms whose excited state couples less strongly to the conduction electrons of the excited state; 3. The use of non-metallic substrates; 4. The development of equipment with increased detection sensitivity. For avenue 1, we are moving towards use of alkali metal substrates whose bulk spectroscopy reveals electronic couplings that become smaller with decreasing Fermi energy E_F . Au already represents an optimal choice among more conventional substrates. Among alternative adsorbates we are considering halogen and chalcogen layers whose optical response on bulk impurities promises lesser quasi-particle-pair creation on the surface also. Studies of H are deferred to situations where its lesser integrated oscillator strength can be accommodated. The third possibility - that of avoiding the conduction electron complication by the use of non-metallic substrates - offers an

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easily achieved spectroscopy but has not yet been pursued strongly as it appears both less interesting and less utilitarian. The fourth development is already fully undertaken as it offers the possibility of studying all adsorbate substrate systems. That is to seek greater equipment sensitivity. The equipment is presently functioning in an essentially ideal manner with a noise figure close to unity and a 100 mc counting limitation. We know of no more accurate equipment for absolute studies of reflection in this energy range. Nevertheless, an improvement by one order of magnitude in sensitivity seems easily within our grasp, and that could well open up the entire adsorbate spectroscopy on metal surfaces to quantitative study.

The sensitivity limitation of $\sim 10^{-3}$ in absolute reflectivity change available from 5-12 eV with our present equipment arises from the photon source, which limits our photon counting times to $\sim 10^7$ photons. During the next few months we will take the equipment, somewhat modified, up to the synchrotron storage ring at Stoughton, Wisconsin, where photon fluxes up to 10^9 /sec are accessible in this energy range and beyond. We anticipate that a sensitivity better than 10^{-4} will be available there, and that quantitative studies of the surface excitations themselves will thus become accessible.

To this end, Dr. Erskine has been interacting with experimental groups currently working at Stoughton and has made use of the ring in a one-time experiment (also designed for the weaker sources available in our laboratories). Drs. Flynn and Erskine, together with Mr. Cunningham, recently attended the Stoughton Users Review Meeting (Oct. 25-27, 1976) to familiarize themselves with the techniques and available facilities. We hope to report the completion of surface measurements using this strong photon source in the near future.

3. Photodesorption. After informative discussions with ONR's Dr. J. Murday we decided that photodesorption experiments could reveal details of atom-surface binding curves, in addition to possessing obvious practical interest in ultra-high vacuum applications. The only quantitatively studied adsorbed system is presently CO, for which the data cover a limited range and have not been explained quantitatively.

Using mainly surplus equipment and an excellent mass spectrometer on permanent loan from Professor A. B. Kunz, we have built up an apparatus for investigating photodesorption with a sensitivity of 10^{-12} atoms ejected per incident photon. This has been achieved by chopping a resonance tube light beam ($\sim 10^{12}$ photons/sec) with synchronous particle detection from the surface by the mass spectrometer operating in the particle-counting mode. A graduate assistant, Mr. R. Moore, helping with this work for his Ph.D., has had his financial needs met by a Physics Department Fellowship. We expect this system to be fully operational, using laboratory photon sources up to 12 eV, in the near future. Further studies at higher photon energies are contemplated for the future, possibly making use of storage ring radiation.

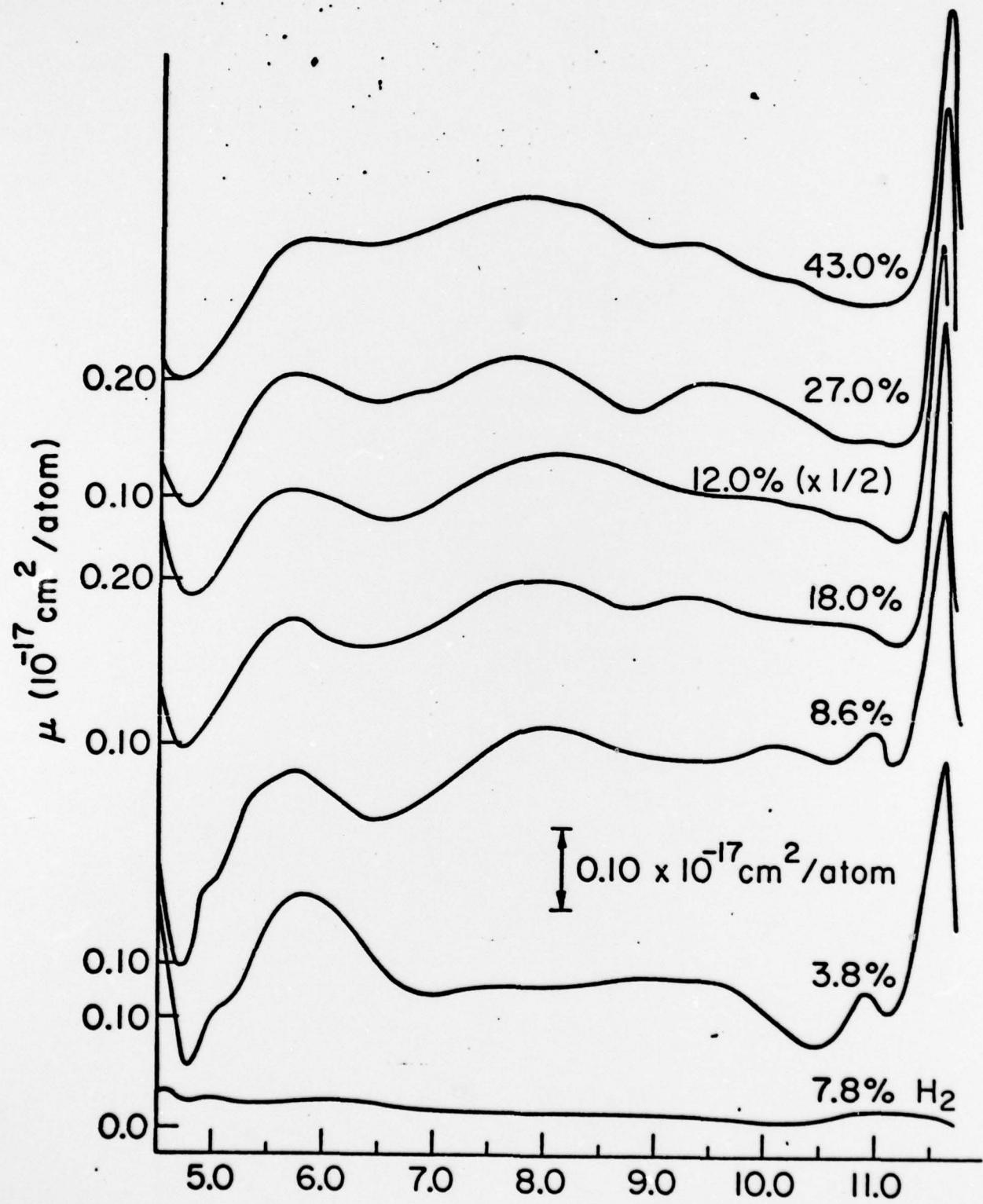
Figure Captions

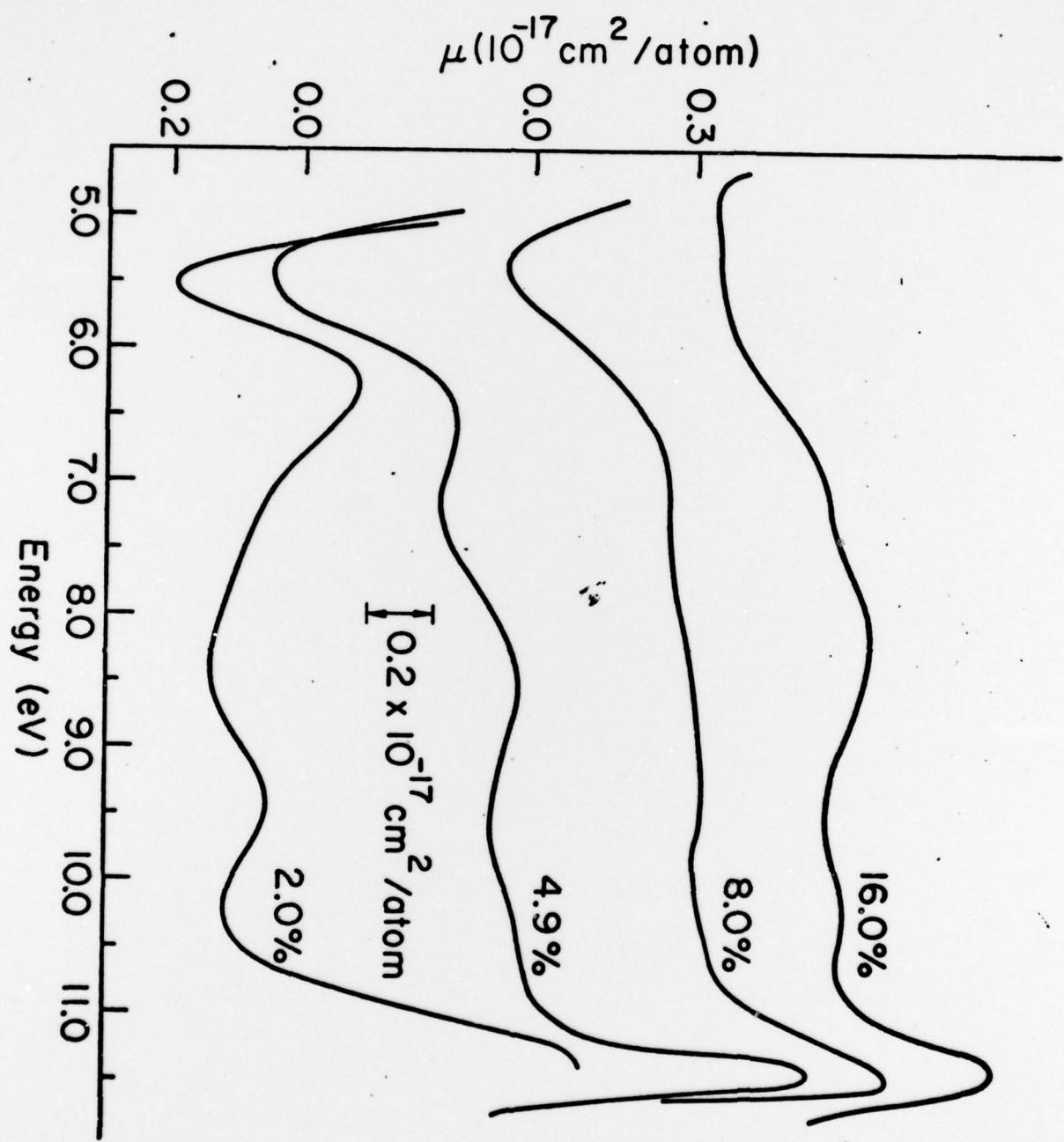
Fig. 1. Atomic hydrogen spectra in Cs thin films. The sharp peak at 11.6 eV identifies the H as interstitially located. The curves show absorption per impurity for various impurity concentrations.

Fig. 2. Oxygen spectra in Cs. Note the 11.5 eV interstitial peak.

Fig. 3. Optical spectra for Xe on Au surfaces (thicknesses given in monolayers).

Fig. 4. Optical spectra for Xe on Ti surfaces (thicknesses given in monolayers).





Xe on Au

